

## Claims

- [c1] CLAIMS:
1. A curable composition, comprising:
    - (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
    - (b) a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
    - (c) a thermoplastic resin; and
    - (d) a cyanate ester.
- [c2] 2. The curable composition of claim 1, wherein said flame retardant additive has a bromine content greater than 20%.
- [c3] 3. The curable composition of claim 1, wherein said flame retardant additive is 1,3,5-tris(2,4,6-tribromophenoxy)triazine.
- [c4] 4. The curable composition of claim 1, wherein said flame retardant additive is 2,2'-[ $(1\text{-methyl} \text{ethylidene})\text{bis}[(2,6\text{-dibromo-4,1-phenylene})\text{oxy}]$ ]bis[4,6-bis [ $(2,4,6\text{-tribromophenyl})\text{oxy}$ ]-1,3,5-triazine].
- [c5] 5. The curable composition of claim 1, wherein said flame retardant additive is soluble in toluene at a concentration of greater than 15 g/100ml of toluene at a temperature of 50 ° C.
- [c6] 6. The curable composition of claim 1, wherein said epoxy resin is a glycidyl ether resin or a mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule.
- [c7] 7. The curable composition of claim 1, wherein said epoxy resin is a mixture of:
  - (a1) an epoxy resin containing on average less than or equal to 2 glycidyl groups per molecule; and
  - (a2) an epoxy resin containing greater than 2 glycidyl groups per molecule.
- [c8] 8. The curable composition of claim 1, wherein said thermoplastic resin has a

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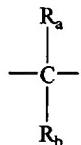
Tg greater than 120 ° C.

- [c9] 9. The curable composition of claim 1, wherein said thermoplastic resin has a dissipation factor of less than 0.010 measured at 1 MHz at room temperature.
- [c10] 10. The curable composition of claim 1, wherein said thermoplastic resin has been directly isolated from solution after polymerization.
- [c11] 11. The curable composition of claim 1, wherein said thermoplastic resin is a poly(phenylene ether).
- [c12] 12. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
- [c13] 13. The curable composition of claim 11, wherein said poly(phenylene ether) has a weight average molecular weight ranging from about 3,000 to 35,000 g/mol.
- [c14] 14. The curable composition of claim 11, wherein said poly(phenylene ether) has been melt processed at a temperature ranging from about 200 ° to 350 ° C.
- [c15] 15. The curable composition of claim 11, wherein said poly(phenylene ether) is hydroxy functional.
- [c16] 16. The curable composition of claim 1, wherein said thermoplastic resin is one or more of a poly(phenylene ether) or a poly(styrene- *co* -maleic anhydride).
- [c17] 17. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether) and a peroxide.
- [c18] 18. The curable composition of claim 1, wherein said thermoplastic resin is a reaction product of a poly(phenylene ether), a peroxide, and a bisphenol.
- [c19] 19. The curable composition of claim 1, wherein said thermoplastic resin is a polyimide.
- [c20] 20. The curable composition of claim 1, wherein the curable composition further comprises one or more of an organic reinforcement, an inorganic

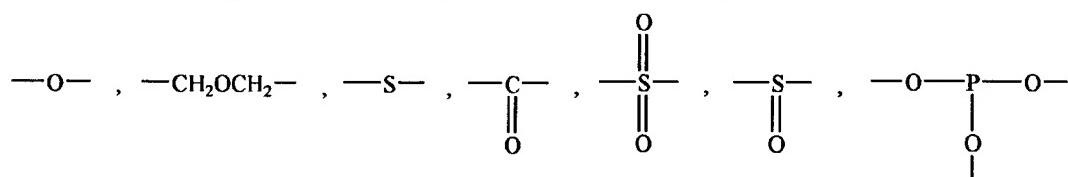
reinforcement, or a filler.

- [c21] 21. The curable composition of claim 1, wherein the curable composition is essentially free of homopolymers of styrene.
- [c22] 22. The curable composition of claim 1, wherein the epoxy resin is a multifunctional glycidyl ether.
- [c23] 23. The curable composition of claim 22, wherein said multifunctional glycidyl ether is selected from the group consisting of epoxidized phenol-formaldehyde novolacs, epoxidized cresol-formaldehyde novolacs, epoxidized alkylphenol-formaldehyde novolacs, epoxidized 1,1,1-tris(4-hydroxyphenyl)ethane, epoxidized 1,1,2,2-tetra(4-hydroxyphenyl) ethane, epoxidized phenol-dicyclopentadiene novolacs, and epoxidized phenol-benzaldehyde novolacs.
- [c24] 24. The curable composition of claim 1, wherein the cyanate ester is selected from the group consisting of 1,3-dicyanatobenzene, 1,4-dicyanatobenzene, 2-tert-butyl-1,4-dicyanatobenzene, 2,4-dimethyl-1,3-dicyanatobenzene, 2,5-di-tert-butyl-1,4-dicyanatobenzene, tetramethyl-1,4-dicyanatobenzene, 4-chloro-1,3-dicyanatobenzene, 1,3,5-tricyanatobenzene, 2,2'-dicyanatobiphenyl, 4,4'-dicyanatobiphenyl, 3,3',5,5'-tetramethyl-4,4'-dicyanatobiphenyl, 1,3-dicyanatonaphthalene, 1,4-dicyanatonaphthalene, 1,5-dicyanatonaphthalene, 1,6-dicyanatonaphthalene, 1,8-dicyanatonaphthalene, 2,6-dicyanatonaphthalene, 2,7-dicyanatonaphthalene, 1,3,6-tricyanatonaphthalene, bis(4-cyanatophenyl)methane, bis(3-chloro-4-cyanatophenyl)methane, bis(3,5-dimethyl-4-cyanatophenyl)methane, 1,1-bis(4-cyanatophenyl)ethane, 2,2-bis(4-cyanatophenyl)propane, 2,2-bis(3,3-dibromo-4-cyanatophenyl)propane, 2,2-bis(4-cyanatophenyl)-1,1,1,3,3,3-hexafluoropropane, bis(4-cyanatophenyl)ester, bis(4-cyanatophenoxy)benzene, bis(4-cyanatophenyl)ketone, bis(4-cyanatophenyl)thioether, bis(4-cyanatophenyl)sulfone, tris(4-cyanatophenyl)phosphate, and tris(4-cyanatophenyl)phosphite.
- [c25] 25. The curable composition of claim 1, wherein the cyanate ester has the formula  
$$R-\left(O-C\equiv N\right)_n$$

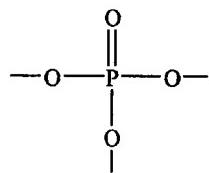
wherein R is an aromatic nucleus-containing residue which is selected from the group consisting of a residue derived from an aromatic hydrocarbon selected from the group consisting of benzene, biphenyl and naphthalene, a residue derived from a compound in which at least two benzene rings are bonded to each other by a bridging member selected from the group consisting of



wherein R<sub>a</sub> and R<sub>b</sub> are the same or different and each represents a hydrogen atom or an alkyl group containing 1 to 4 carbon atoms,



and

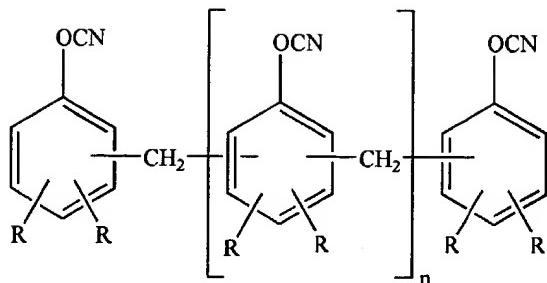


and a residue resulting from the removal of a phenolic hydroxyl group from a novolac-type or resol-type phenolic resin skeleton; said aromatic nucleus is optionally substituted by a substituent selected from the group consisting of alkyl groups containing 1 to 4 carbon atoms, alkoxy groups containing 1 to 4 carbon atoms, chlorine and bromine; n is an integer of 2 to 5; and the cyanate group is always directly bonded to the aromatic nucleus.

- [c26] 26. The curable composition of claim 1, wherein the cyanate ester is a prepolymer of the cyanates esters of Claim 25, having a number average molecular weight of 400 to 6,000, and are formed by trimerizing the cyanate group of the cyanate esters.

[c27]

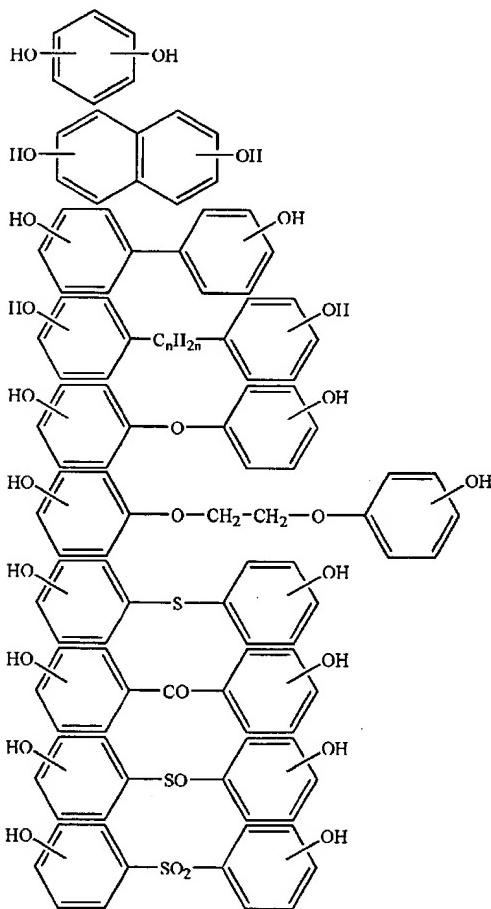
27. The curable composition of claim 1, wherein the cyanate ester is a cyanate-group-containing phenol resin comprising a mixture of polymers represented by the formula



wherein n is 0 or an integer of 1 or more; and R's may be the same or different, and each R is a hydrogen atom or a methyl group, and containing 50% by weight or more in total of polymers having formula in which n is an integer of 1 to 3, the number average molecular weight of said phenol resin being 350 to 700 g/mole.

[c28]

28. The curable composition of claim 1, wherein the cyanate ester is a cyanic acid ester of an aromatic polycarbonate obtained by reacting an aromatic polycarbonate having one or two terminal hydroxyl groups with a cyanogen halide, wherein the aromatic polycarbonate is prepared from an aromatic dihydroxy compound represented by one of the following formulas:



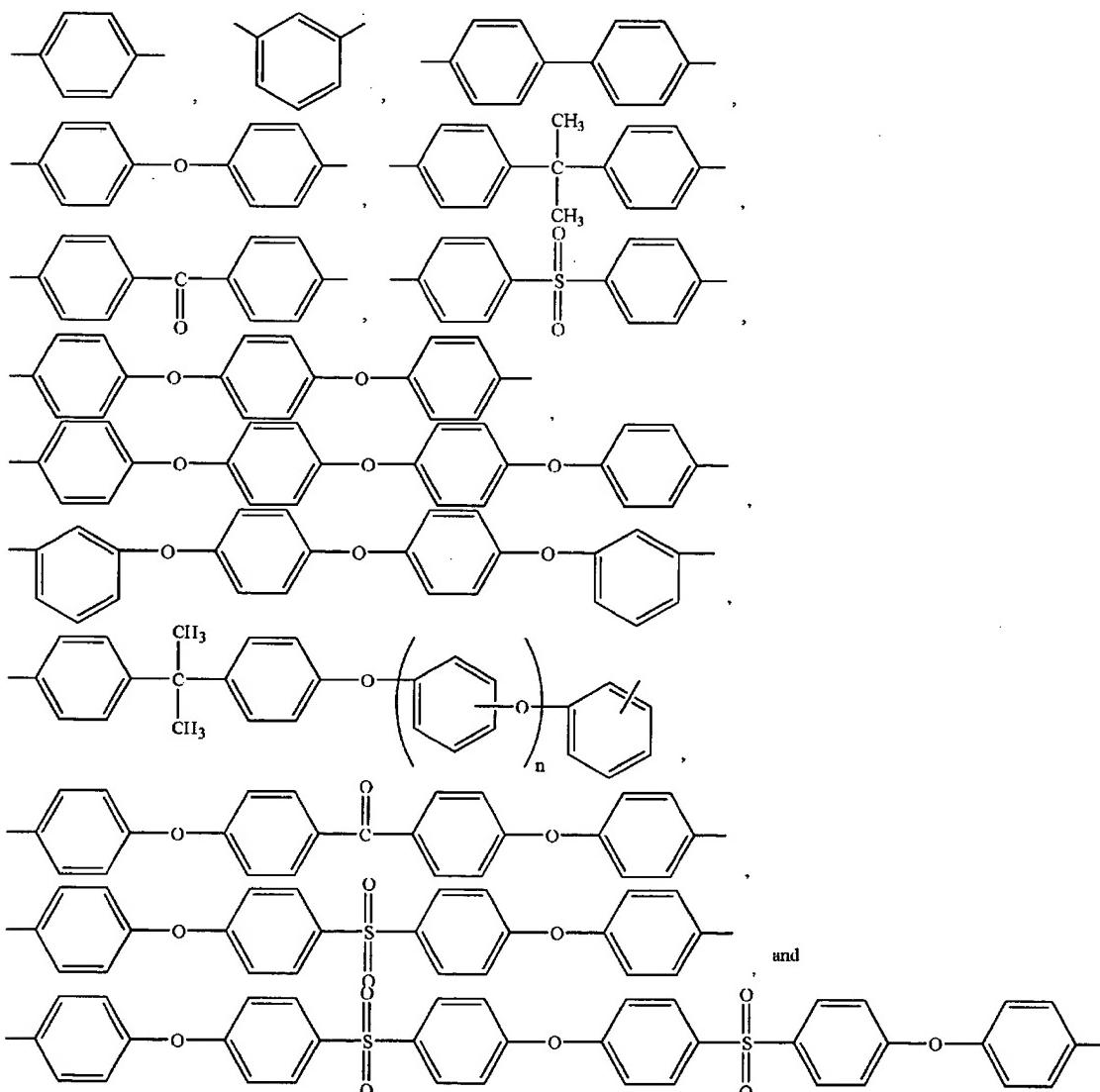
where n is an integer of 1-4, inclusive, or a mono-, di-, tri- or tetra- halogeno-

nuclear substituted derivative of the aromatic dihydroxy compound represented by one of the above formulas.

- [c29] 29. The curable composition of claim 1, wherein the cyanate ester is a cyanatophenyl-terminated polyarylene ether of the formula

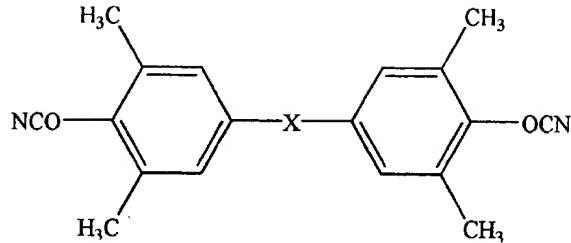


where R is a divalent radical having 3 to 15 aromatic nuclei linearly linked together with ethereal oxygen atoms, said nuclei comprising nuclei selected from the group consisting of



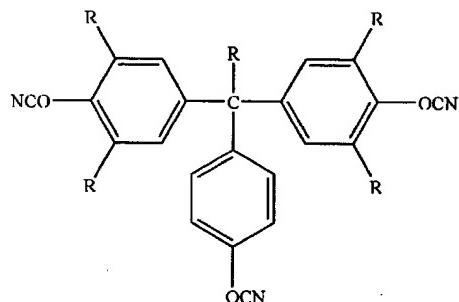
- [c30]

30. The curable composition of claim 1, wherein the cyanate ester has the structure

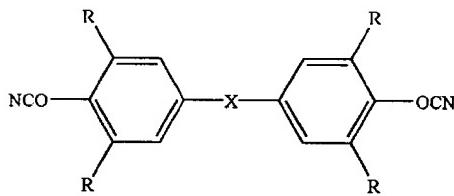


wherein X is methylene, isopropylidene, oxygen or divalent sulfur.

- [c31] 31. The curable composition of claim 1, wherein the cyanate ester is a blend of a tricyanate ester and a dicyanate ester, wherein the tricyanate ester has the structural formula:



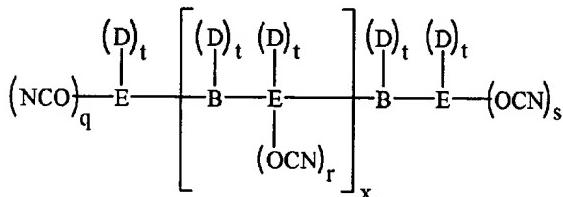
and the dicyanate ester has the structural formula:



wherein each R is H or methyl and is the same or different and wherein X is methylene, alkylidene having 2 to 4 carbon atoms, divalent oxygen, or divalent sulfur.

- [c32]

32. The curable composition of claim 1, wherein the cyanate ester is a polyaromatic cyanate having the formula



wherein: E is an aromatic radical;

B is a  $C_{7-20}$  polycyclic aliphatic radical;

D is independently in each occurrence any nonactive hydrogen-containing substituent;

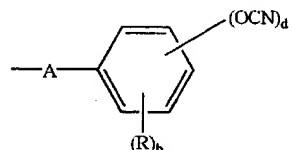
q, r and s are independently in each occurrence the integers 0, 1, 2, or 3; with

the proviso that the sum of q, r and s is greater than or equal to 2; t is independently in each occurrence an integer of between about 0 and 4 inclusive; and x is a number between about 0 and 5 inclusive.

- [c33] 33. The curable composition of claim 1, wherein the cyanate ester is a fluorocarbon monocyanate having the structure  
 $F_3C(CFX)_aA(CFX)_bCH_2OCN$   
where A is  
 $\left( O-CFX-CF_2-O \right)_c$ ,  
X is fluorine or perfluoroalkyl having 1 to 10 carbon atoms, a is 1 to 10, h is 1, and c is 1 to 100.
- [c34] 34. The curable composition of claim 1, wherein the cyanate ester is a fluorocarbon dicyanate having the structure  
 $NCOCH_2(CFX)_aB(CFX)_bCH_2OCN$   
where B is (I) a carbon-to-carbon bond, in which case a is an integer of 1 to 30 and b is zero, or (II) B is  $[(CFX)_dO(CFX)_e]_f$ , in which case a and b are zero, d and e are integers of 1 to 30, and f is an integer of 1 to 20, or (III) B is  $(OCF_2-CFX)_gO(CFX)_hO(CFX-CF_2O)_i$ ,  
in which case a and b are 1, h is an integer of 1 to 10, and g and i are integers of 1 to 100, or (IV) B is  
 $[(CF_2CH_2)_j(CFX-CFX)_k]_m$ ,  
in which case a and b are integers of 1 to 10, j and k are integers whose ratio j/k is 1/1 to 10/1, m is an integer of 1 to 100, and  $(CF_2CH_2)$  and  $(CF_2-CFX)$  are randomly distributed units; and where X in all instances where it appears is fluorine or perfluoroalkyl of 1 to 10 carbon atoms.
- [c35] 35. The curable composition of claim 1, wherein the cyanate ester has the formula
- 
- in which R represents hydrogen, halogen, linear or branched C<sub>1</sub>-C<sub>9</sub>-alkyl or phenyl, two adjacent radicals R on the same nucleus together forming a

carbocyclic 5-membered or 6-membered ring or together and in conjunction with a hetero atom (O, S, N) forming a 5-membered or 6-membered heterocyclic ring, alkoxy radicals with 1 to 4 carbon atoms, alkoxy carbonyl radicals with 1 to 4 carbon atoms in the alkyl group;

R' has the same meaning as R or represents the group



where A is direct bond, a C<sub>1</sub>–C<sub>9</sub>-alkylene group optionally substituted by C<sub>1</sub>–C<sub>4</sub>-alkyl or phenyl, a cycloaliphatic or aromatic 5-membered or 6-membered ring, or a cycloaliphatic or aromatic 5-membered or 6-membered ring; a is a number from 0 to 5 where e = 1 and a number from 2 to 5 where e = 0; b = 5 – a where e = 1 and 6 – (a + d) where e = 0; c = 5 – d; d is a number from 0 to 5; e is the number 0, 1, 2 or 3; with the proviso that the sum of a and d (a + d) always gives a number from 2 to 5.

- [c36] 36. A curable composition, comprising:
- (a) an epoxy resin and curing agent therefor, wherein said epoxy resin is a glycidyl ether resin or mixture of glycidyl ether resins containing, on average, greater than 2 epoxy groups per molecule;
  - (b) 1,3,5-tris(2,4,6-tribromophenoxy)triazine and/or 2,2'-[(1-methylethylidene)bis[(2,6-dibromo-4,1-phenylene)oxy]]bis[4,6-bis[(2,4,6-tribromophenyl)oxy]-1,3,5-triazine];
  - (c) a poly(phenylene ether) resin; and
  - (d) a cyanate ester.
- [c37] 37. A curable composition, comprising:
- (a) an epoxidized cresol-formaldehyde novolac resin;
  - (b) 1,3,5-tris(2,4,6-tribromophenoxy)triazine;
  - (c) a poly(phenylene ether) resin having a number average molecular weight ranging from about 1,000 to 15,000 g/mol; and
  - (d) a cyanate ester.
- [c38] 38. A cured composition comprising a cured residue of a curable composition comprising:

- (a)an epoxy resin and curing agent therefor, wherein said epoxy resin is essentially free of bromine atoms;
- (b)a flame retardant additive essentially free of phenolic groups and of epoxy groups, wherein said flame retardant is a condensation product of (i) a brominated phenol or a mixture of brominated phenols with (ii) a cyanuric halide;
- (c)a thermoplastic resin; and
- (d) a cyanate ester.